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ELECTROSORPTION OF METAL IONS FROM AQUEOUS SOLUTIONS

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ELECTROSORPTION OF METAL IONS FROM AQUEOUS SOLUTIONS

INTRODUCTION

Electrosorption has been investigated for the purpose of heavy metal removal and desalination for a long time. Although electrosorption has been shown as a promising treatment process, it has been limited by the performance of electrode material, specifically, the electrode lifetime. Recently, carbon aerogel—a monolithic, high surface area, and high electrical conductivity material—has been developed at National Lawrence Livermore Laboratory (Mayer et al., 1993). Electrosorption by using carbon aerogel electrodes has several advantages over other conventional technologies. Unlike ion exchange, no acids, bases, or salt solutions are required for regeneration of the system. Therefore, the amount of secondary waste is largely reduced. In contrast to thermal processes such as evaporation, electrosorption does not consume much energy and thus increases energy efficiency. Electrosorption also has operational advantages over electrodialysis and reverse osmosis because no separation membranes are required.

Several researchers have investigated the use of porous electrodes for the recovery of heavy metals from aqueous solutions (Johnson and Newman, 1971; Matlosz and Newman, 1986). In these cases, metallic ions are electrodeposited on the surfaces of cathodes. The problem with electrodeposition is that the regeneration of the electrodes is relatively difficult. The idea of electrosorption is, therefore, introduced as an alternative process. Electrosorption is generally defined as potential-induced adsorption on the surface of electrodes. More precisely, electrosorption by porous electrodes can be viewed as an adsorption process that takes place in an electrochemical double-layer capacitor. After polarization of the electrodes, charged impurities are transported to the electrode surfaces and held within the electrical double layers to produce an effluent of purified water. Once discharging the electrodes, the ions will be quickly released back to the bulk solution. This reversible behavior largely increases the regeneration efficiency and reduces the secondary waste. Furthermore, since acting like an electrical capacitor, the electrosorption process does not consume much energy and thus lowers the operating cost (Mayer et al., 1993; Bispo-Fonseca et al., 1999).

Although all materials exhibit double-layer capacitance, there are four important requirements for electrodes used in such applications: (1) high surface area, (2) low electrical resistance, (3) good polarizability, and (4) no participation in faradaic reactions at the applied voltage (Pekala, 1998). In the earlier studies of electrosorption (Koresh and Soffer, 1977; Koresh and Soffer, 1983; Oren et al., 1978; Oren et al., 1984; Golub et al., 1989; Trainham and Newman, 1977; Risch and Newman, 1984), researchers have shown good removal efficiency of metal ions by activated carbon beds. Several practical problems are, however, encountered with activated carbon electrodes. For example, significant fractions of the activated carbon surface may be occluded in electrodes that

use polymeric binders (Arnold and Murphy, 1961). Such electrodes have characteristically high electrical and mass-transfer resistance. In addition, polymer binders are susceptible to both chemical attack and radiation-induced degradation.

Recently, carbon aerogel—a type of carbon foam—has been developed by Mayer et al. (1993) to be used as an electrical double-layer capacitor for energy storage applications. The carbon aerogel electrodes are highly porous and monolithic, with high surface area ($\sim 400 - 1000 \text{ m}^2/\text{g}$), low electrical resistivity ($\leq 40 \text{ m}\Omega \text{ cm}$), and controllable pore size distribution ($\leq 50 \text{ nm}$) (Mayer et al., 1993; Wang et al., 1993). Carbon aerogels mainly contain two pore size regimes: (a) micropores ($< 2 \text{ nm}$) and (b) mesopores ($2 - 50 \text{ nm}$). Farmer et al. (1996; 1997) have conducted experiments for the removal of ions such as sodium chloride, chromium ions, and ammonium perchlorate by electrosorption using carbon aerogel electrodes. They claimed that although double-layer charging is the major mechanism, chemisorption also plays a significant role for certain ions, e.g., chromium (Farmer et al, 1997).

Oren et al. (1984; 1985; 1986) have investigated the electrical double-layer capacitance on carbon and graphite over a wide potential range by monitoring the dimensional changes on the electrode surface. They found that during the charging process on carbon or graphite electrodes, faradaic or electrochemical charge that crossed the interface is significant at high voltage. Wang et al. (1993), however, showed from their experimental measurements that the possibility of faradaic reactions on the carbon aerogel electrodes is very small compared to that on carbon or graphite. This advantage combined with high surface area and low electrical resistivity makes carbon aerogel composites an almost ideal electrode material for electrosorption.

There are two main mechanisms that have been suggested for the electrosorption process on porous electrodes: electrical double layer formation and chemical reactions on electrode surfaces. To simplify the simulation system, earlier modeling studies usually considered only electrical double layer formation. Johnson and Newman (1971) developed a comprehensive theoretical model for ionic adsorption on porous carbon electrodes. Their analysis basically simulates the electrosorption process from a macroscopic view and shows a good agreement with experimental data. Farmer et al. (1996; 1997) attempted to use the Gouy-Chapman theory, which is developed for simple planar electrodes, to explain their experimental results on carbon aerogel electrodes. Their modeling results, however, show that the surface charge density fails to meet a square root dependence on electrolyte concentration, which may be due to the self-shielding effect experienced by the porous carbon electrodes. Since carbon aerogel composites have a high fraction of micropores, the self-shielding or double-layer overlapping effect inside the porous electrodes becomes significant and needs to be considered for the simulation of electrosorption processes from a microscopic view.

By charging porous electrodes, electrical double-layers are formed inside the electrode pores, instead of adjacent to the electrode surface. The pore structure greatly increases the effective area of the electrode as well as the electrical capacity. However,

when the pore size is of the same order of magnitude as the thickness of the electrical double-layer, the electrical double-layers inside the pore will overlap onto each other and lose their electrical capacities. Since this overlapping effect mainly exists in microporous (< 2 nm) and part of mesoporous (2-50 nm) regions, most of the electrical double-layer models do not take it into consideration. For carbon aerogel electrodes, the pore size is generally smaller than 100 nm; therefore, it is necessary to include the overlapping effect for the prediction of the electrical capacities. Besides the overlapping effect, the repulsive ion-dipole interaction of the ions with the dipole of the chemisorbed oxygen can be important inside the porous structure.

In the present work, the results of bench-scale equilibrium experiments of electrosorption by using carbon aerogel to remove various ions are reported. The effects of various parameters such as solution concentration, pH, and applied voltage on the electrosorption performance are discussed. The pore size distribution of the carbon aerogel is also examined in this work. In addition, an electrical double-layer model that includes the overlapping effect is developed to predict the electrosorption capacity on carbon aerogel electrodes. In order to focus on the electrical double-layer formation, the model does not consider specific adsorption at this point. Using the pore size distribution, the model predicts the effects of applied voltage and solution concentration on the electrosorption capacity and cut-off diameter. The cut-off diameter is defined as the smallest pore size in which the electrical double-layer can be formed. When the overlapping effect is incorporated in the model, it is demonstrated that modeling results agree well with the experimental data.

MATERIALS AND METHODS

The major objective of this project is to investigate ion removal from aqueous solutions by electrosorption using carbon aerogel electrodes. The focus is on understanding the mechanisms and quantifying the effects of important parameters on the equilibrium capacity of ion removal by electrosorption.

Characterization of Carbon Aerogel. Pore size distribution and electrical capacitance are the two most important properties needed to be characterized for carbon aerogel. Pore size distribution determines the available surface area for electrosorption; on the other hand, electrical capacitance determines the electrosorption capacity per unit area of carbon aerogel. Experimental studies for the characterization of carbon aerogels include surface charge density, electrical capillary maximum (ECM), and pore size distribution measurements.

The surface charge density of carbon aerogel material was measured by using a potentiometric titration method with a Metrohm automatic titration system. The solution was prepared by adding ground carbon aerogel into deionized water, and the desired ionic strength was adjusted by using NaNO_3 . During the titration, the pH was adjusted by using 0.1 M HNO_3 or 0.1 M NaOH .

ECM measurements were conducted by continuously increasing the applied voltage at a constant rate (0.33 mV s^{-1}) and recording the current change between a pair of carbon aerogel electrodes. The region where current did not change with voltage gives the ECM of carbon aerogel. The ECM experiments were conducted at various concentrations of NaF solution from 25 ppm to 300 ppm. The reason for choosing NaF solution is because fluoride ions do not react with the carbon aerogel surface; thus, the simulation system is simplified since ions can be treated as point charges.

The pore size distribution and specific surface area of the carbon aerogel were measured by using physical adsorption and mercury intrusion porosimetry. These characterization experiments were conducted by Dr. E.S. Vittoratos at Chevron Research and Technology Company.

The physical adsorption measurements were conducted by a Micromeritics ASAP 2010 instrument, using N_2 at 77 K and CO_2 at 273 K. All gases were of at least 99.99% purity. The carbon aerogel sample was prepared by outgassing it for 16 h at 300°C under vacuum; upon transfer to the analysis port of the instrument, it was heated for an additional 30 min at the same temperature under vacuum. The sample was then permitted to cool under vacuum prior to the commencement of the adsorption measurements. Physical adsorption with N_2 gas was conducted. Initially, N_2 gas was added in one or two cm^3 increments at standard temperature and pressure (STP), and the pressure in the sample tube was permitted to equilibrate. The same procedure was continued until a partial pressure of 0.14 was reached; then, sufficient gas was admitted to achieve increasing pre-defined partial pressures up to 0.99. Physical adsorption with CO_2 gas followed a similar procedure but began with smaller gas increments: about 0.1 cm^3 STP doses, until a partial pressure of 0.03 was achieved. Carbon dioxide physical adsorption was found to be an accurate method for characterizing pores of sizes smaller than 10 \AA (Garrido et al., 1987). Physical adsorption results were then analyzed using density functional theory to provide the pore size distribution of the material (Lastoskie et al., 1993; Olivier, 1995).

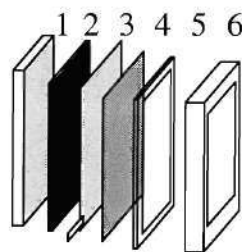
Mercury intrusion porosimetry was performed with a Quantachrome instrument. The pressure was continuously increased from one atm to 4,000 atm in ten minutes. Mercury porosimetry needs to be used with caution for soft materials such as carbons or silica because high pressures can potentially compact the samples (Vittoratos and Auburn, 1995).

Equilibrium Experiments of Electrosorption. The removal of dissolved inorganic contaminants, such as radionuclides, metal ions, and anions is a widespread need for groundwater and wastewater treatment. Electrosorption is a promising technology for application to these needs that involve streams with low-to-moderate ionic strength (i.e., it is not applicable to high-salt tank wastes); it may also be an improvement over existing treatment systems on other streams. The experimental work of this project is expected to provide a better understanding of the performance and mechanisms of electrosorption by

using carbon aerogel electrodes, and thereby improve adsorbent capacity and treatment performance.

Batch experiments were conducted by using a pair of carbon aerogel electrodes. As shown in Figure 1, the electrode was made by attaching one carbon aerogel composite [indicated as (4)] onto a titanium plate [indicated as (3)] that serve as both a current collector and a structural support for the electrodes. Each sheet of carbon aerogel has dimensions of $15.6 \times 8 \times 0.0127 \text{ cm}^3$. The reaction cell was assembled within two plexiglass plates [indicated as (1) in Figure 1]. The separation distance between the electrodes was maintained at 0.6 cm by a plexiglass spacer [indicated as (6) in Figure 1] that has a hole of $15.1 \times 7.6 \text{ cm}^2$ at its center.

In each experiment, the solution was continuously pumped from a recycling reservoir into the cell and the effluent returned back to the reservoir. The electrodes were connected to a (direct-current) dc power supply with a voltage range from 0 to 15 V and a current range of 0 to 7 A. Solution pH, conductivity, temperature, and current were continuously monitored. During the experiment, the solution was run through the cell without an applied voltage until the system reached equilibrium. A desired voltage was then applied between the electrodes until a new equilibrium was reached. Regeneration was accomplished by discharging the cells at 0 V, or by reverse polarization at an opposite charge. Reverse polarization can increase the regeneration effectiveness and/or reactivate the carbon aerogel electrodes. Most of the experiments were using sodium fluoride and sodium chloride solutions. The reason for choosing fluoride and chloride as target ions is because the chemisorption for these two chemical species on carbon aerogels is insignificant and thus can simplify our modeling system. Different solutions such as NaBr, NaI, and $\text{Cu}(\text{NO}_3)_2$ were also used to study the effects of anion type and multivalent metal ions on electrosorption.



(1) Plexiglass cover (4) Carbon aerogel
(2) Viton gasket (5) Viton gasket w/ hole
(3) Titanium plate (6) Central plexiglass

Figure 1. Assembly of the electrosorption cell (half of the cell is shown above).

RESULTS AND DISCUSSION

A. Experimental Results

Surface Charge Density. Figure 2 shows the results of surface charge density measurements, obtained from potentiometric titrations. The point where the three lines of various ionic strengths are crossed at is defined as the point of zero charge. From Fig. 2, the point of zero charge (pzc) is at pH 9.2, and the surface charge increases with

decreasing pH. The surface charge density found on the carbon aerogel surface is very small and is comparable to that on activated carbon material (Corapcioglu and Huang, 1987; Chen et al., 1997). This result indicates that the electrostatic force is insignificant when there is no external potential applied.

ECM Measurements. Experiments for measuring ECM were conducted at different sodium fluoride concentrations from 25 ppm to 300 ppm. It was found that the constant current range happened at applied voltage of around 3.6 V, which gives an ECM value of around 0.18 V.

Pore Size Distribution. Pore size distribution data of carbon aerogel were obtained by superimposing the results of two gas adsorption methods (CO_2 for micropores and N_2 for mesopores) and then analyzing the results by density functional theory (DFT). The pore size distribution of carbon aerogel is shown in Fig. 3. These results show that there are three peaks in the pore size distribution: they are around 8 Å, 20 Å, and 100 Å, respectively. Also, there are very few pores with a size greater than 1000 Å. Meanwhile, a great portion of pore sizes falls below 10 Å, where strong electrical double layer overlapping is expected to happen.

Effect of Anion Species. The effect of type of anion species on the electrosorption capacity at different concentrations is shown in Fig. 4. In each set of columns, the first bar represents the chemisorption capacity and the second one represents the electrosorption capacity. The chemisorption capacity is defined as the amount of ion removal without an applied voltage. On the other hand, the electrosorption capacity is defined as the amount of ion removal with an applied voltage. From the experimental results, it was found that the chemisorption capacity is relatively small compared to the electrosorption capacity. Also, the capacity of anion removal increases in the order of Cl^- , Br^- , and I^- . The reason is probably because the iodide ions have a higher partial charge-transfer coefficient than the bromide and chloride ions. The partial charge-transfer coefficient is an indicator of how many electrons can be released from an adsorbate to an adsorbent during adsorption. In addition, it was observed that at higher concentrations, the electrosorption capacity is increased. Since sodium ions are the only cations shown in the system, it can be concluded that the electrosorption capacity is limited by anions.

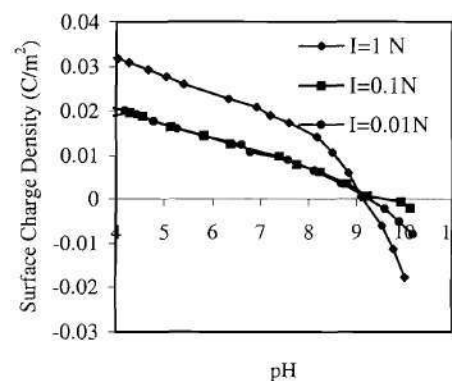


Figure 2. Surface charge density measurements for carbon aerogel.

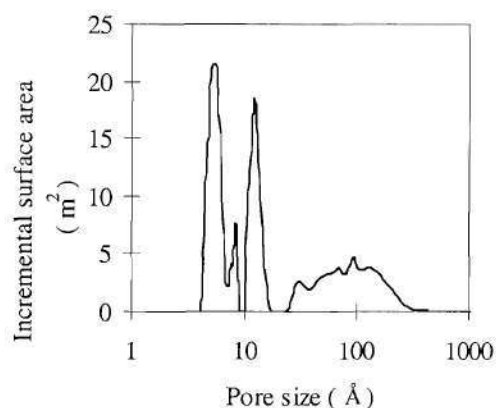


Figure 3. Incremental surface area as a function of pore size for carbon aerogel.

Effect of Solution Concentration. The effect of initial concentration on the deionization capacity of sodium fluoride ions was investigated by varying the solution concentration from 30 ppm to 1000 ppm at an applied voltage of 1.2 V. As shown in Fig. 5, a logarithmic relationship between total capacity and initial concentration was found. The deionization capacity of carbon aerogel was found to increase with an increase of the initial solution concentration. Although the electrosorption capacity increased as the initial concentration was increased, the removal efficiency of NaF dropped from 70% to 15% as the initial concentration was increased from 30 ppm to 1000 ppm. It can be concluded that electrosorption is more efficient in dilute systems. From the experimental results, it was also observed that the electrosorption capacity changed with the age of the carbon aerogel.

Effect of Applied Voltage. The effect of applied voltage on electrosorption capacity is shown in Fig. 6. It is found that the electrosorption capacity did not increase much at low applied voltage (< 0.6 V), and then increased quickly when the applied voltage was increased from 0.6 to 1.3 V. The increasing rate of deionization capacity decreased at high voltage (> 1.3 V), and also bubbles were observed as the applied voltage became higher than 1.2 V. The reason that electrosorption capacity does not increase much as voltage is increased may be due to the electrical capillary maximum (ECM) of the carbon aerogel composite. It was found from experimental results that the value of ECM is around 0.18 V with respect to one electrode. The electrostatic driving force, therefore, will be used to balance the ECM voltage until the applied voltage increases beyond 0.36 V.

Effects of Solution pH and Flow-Rate. During the experiments, it was observed that solution pH increased very quickly from about pH 7 to pH 10 when voltage was applied and slowly dropped back to its initial pH value. This observation implies that when voltage is applied, OH^- ions are released from the electrode surface. One-side carbon

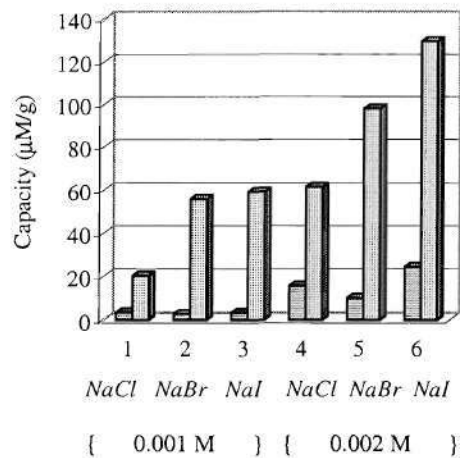


Figure 4. Effect of anion type on the electrosorption capacity; $V=1.2$ V.

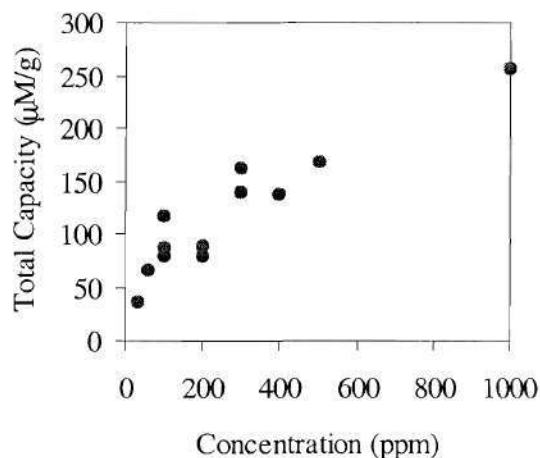


Figure 5. Effect of solution concentration on electrosorption capacity; $V=1.2$ V.

aerogel experiments, therefore, were conducted, in which one of the electrodes (anode or cathode) was made by carbon aerogel and the other by platinum. It was observed that the solution pH did not change much when the carbon aerogel electrode served as cathode; the solution pH, however, increased when the carbon aerogel electrode served as anode. This result indicates that the increase of pH in the experiments of two-side carbon aerogel electrodes is due to anode reactions. In addition, the effect of flow-rate on the equilibrium electrosorption capacity was studied by varying the flow-rate from 8 mL/min to 54 mL/min. The experimental results showed that the difference of equilibrium capacities at different flow-rates was insignificant. It means that the change of flow-rate does not affect the equilibrium capacity of electrosorption.

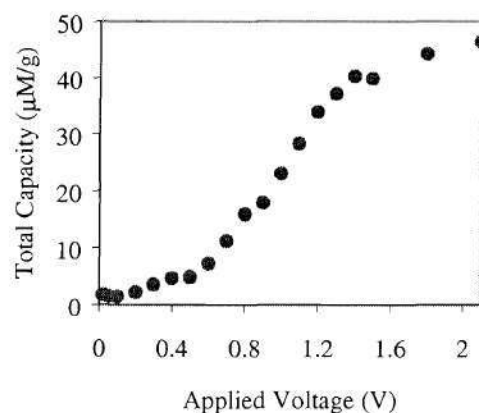


Figure 6. Effect of applied voltage on electrosorption capacity; concentration = 100 ppm NaF.

Removal of Copper Ions. Figure 7 shows experimental results for the removal of copper ions by electrosorption using carbon aerogel electrodes. Significant chemisorption of copper ions occurred and the removal efficient was about 59% without imposing an electric field. After a voltage application of 1.2 V, some of the ions were desorbed back to the solution and then removed by electrosorption to reach a much higher removal efficiency of 93%. In addition, after the electrodes had been discharged, the solution conductivity did not go back to its initial value, which indicates that copper ions form complexes on the carbon aerogel surface. Further studies on the surface characteristics of carbon aerogels need to be conducted to better understand its interaction with ions.

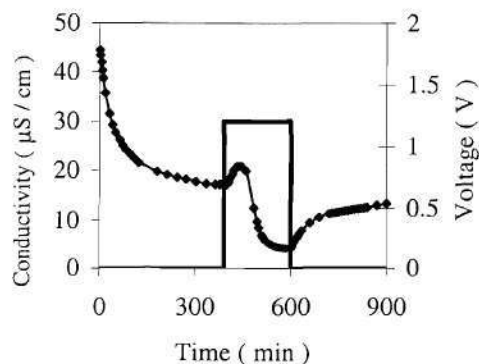


Figure 7. Removal of copper ions by electrosorption on carbon aerogel electrodes; $V = 1.2$ V; concentration = 40 ppm.

B. Model Development

Electrical double layer capacity can be interpreted by the Gouy-Chapman theory on a flat surface. For porous electrodes such as carbon aerogel, the electrical double-layers

form inside the electrode pores instead of adjacent to the electrode surfaces. The pore structure greatly increases the effective area of the porous electrodes as well as the electrical capacity. However, when the pore size is of the same order of magnitude as the electrical double-layer thickness, the electrical double-layers inside the pore will overlap onto each other and lose their electrical capacities. Since this overlapping effect only exists in microporous (< 2 nm) and part of mesoporous (2-50 nm) regions, most of the electrical double-layer models do not take it into consideration. Carbon aerogel composites have a significant fraction of micropores; it is, therefore, necessary to consider the overlapping effect. The cut-off diameter is the smallest pore size in which the electrical double layer can exist, and can be determined by

$$d_m = \frac{2}{-\kappa} \ln \frac{[\exp(e\psi_d/kT) + 1](e\psi_d/kT)}{8[\exp(e\psi_d/2kT) - 1]} \quad (1)$$

where d_m is the cut-off diameter, κ is the Debye-Hückel parameter, e is the elementary charge, k is the Boltzmann constant, T is the absolute temperature, and ψ_d is the diffuse layer potential of the electrical double layer. The cut-off diameter d_m usually ranges from 10 to 100 Å, depending upon the diffuse layer potential and the Debye-Hückel constant κ . Pores with diameter less than the cut-off value will not contribute to the electrosorption of ions. Figure 8 shows the effect of pore size on the total capacity and also the cut-off diameter at different concentrations. It is found that when the pore size is smaller than 40 Å, the total capacity decreases rapidly and reaches zero after a specific pore size. This cut-off diameter decreases as the concentration is increased.

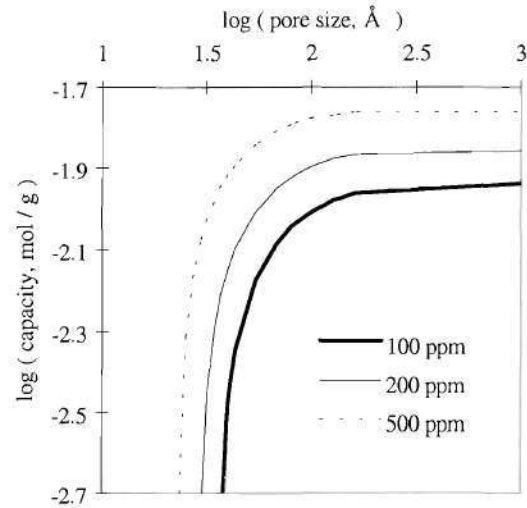


Figure 8. Cut-off diameter at different concentrations; $V=0.6$ V.

An electrical double layer model that considers double-layer overlapping correction has been developed to predict the total capacity of electrosorption on carbon aerogel. The model can be used to predict the total capacity under various values of concentration and applied voltage. In this electrical double layer model, the diffuse layer potential, ψ_d , is one of the most important parameters needed to calculate the electrosorption capacity. For a carbon aerogel electrode under applied voltage V , the diffuse layer potential can be expressed as follows:

$$\psi_d = \frac{V}{2} - \phi_{ecm} - \int_0^{\sigma_0} \frac{1}{C_1(\sigma_0)} d\sigma_0 \quad (2)$$

where ϕ_{ecm} is the electric potential at the electric capillary maximum (ECM), C_1 is the inner layer capacitance, V is the applied voltage, and σ_0 is the surface charge density.

By inserting this equation into the Poisson-Boltzmann equation, the surface charge density can be calculated. The electrosorption capacity is, thus, calculated by integrating the individual capacity in each pore over the entire pore size distribution data. The theoretical charge density can be calculated using the following equation:

$$\sigma_0 = \sqrt{4\epsilon RTI} \left[\cosh \left(\frac{e \left(\frac{V}{2} - \phi_{ecm} - \frac{\sigma_0}{32} \right)}{kT} \right) - \cosh \left(\frac{e\psi_m}{kT} \right) \right]^{1/2} \quad (3)$$

where R is the ideal gas constant, I is the ionic strength, ϵ is the dielectric constant, and ψ_m is the electrical potential at the centerline of the pore. The surface charge density equation can be solved by iteration.

The comparison between modeling results and experimental data is shown in Fig. 9. It is demonstrated that the total capacity increases with applied voltage. It is also found that without including the overlapping effect, the electrosorption capacity (indicated in the figure with thin solid line) will be largely overestimated. On the other hand, by including the overlapping effect (indicated in the figure with thick solid line), a good agreement between experimental data and modeling results is found. The deviations observed at high voltage are attributed to faradaic reactions that may become significant at high voltage and, therefore, decrease the total capacity.

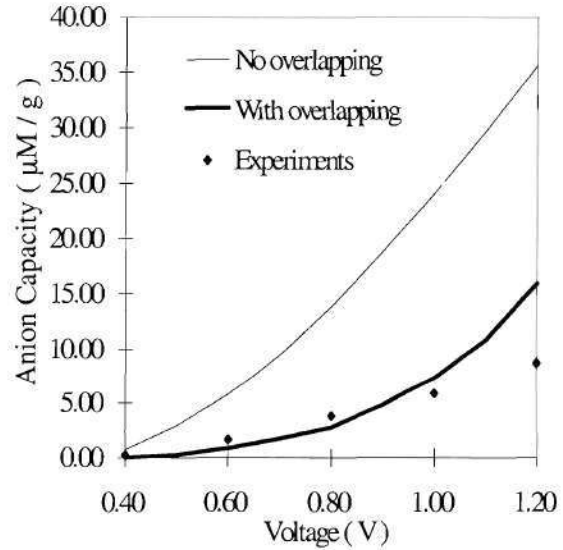


Figure 9. Total anion capacity as a function of applied voltage; concentration=100 ppm NaF.

SUMMARY

Our experimental and modeling studies demonstrate that electrosorption is an ideal process for removing ions from aqueous solutions. The experimental data indicate that

the electrosorption capacity of carbon aerogel increases with an increase of the initial solution concentration and applied voltage. The removal efficiency of anions increases in the order of chloride, bromide, and iodide, which agrees with the order of their corresponding partial charge-transfer coefficients. In addition, it is found that the electrosorption process largely enhances the removal efficiency of copper ions from 59% to 93%.

An electrical double-layer model that includes the effect of electrical double-layer overlapping is developed in this work. The modeling results show that it is necessary to consider the overlapping effect for microporous materials. A good agreement of modeling results with experimental data is obtained.

Electrosorption phenomena investigated in this project can be readily employed to enhance the separation of metal ions and anions from aqueous solutions. Electrosorption by carbon aerogel can be applied in situ, and the carbon aerogel material can be readily regenerated by removing the electric field and redirecting the flow. The basic knowledge obtained here can be used to design the properties of the electrode material so that the electrosorption capacity will be maximized.

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